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OAK RIDGE NATIONAL LABORATORY

NASA TUNGSTEN REACTOR RADIATION CHEMISTRY STUDIES

PHASE I, EXPERIMENT DESIGN

G. H. Jenks
H. C. Savage
E. G. Bohlmann

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NASA TUNGSTEN REACTOR RADIATION CHEMISTRY STUDIES

PHASE I, EXPERIMENT DESIGN

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1. Introduction

Poison control solutions of $CdSO_4$ are being considered for possible use in the NASA Tungsten Water Moderated Reactor. Information regarding effects of irradiation on the stability of these solutions toward loss of cadmium is needed for complete evaluation of this poison control system. We have planned and are developing experiments to test the stability of $CdSO_4$ solutions under electron irradiation (using a Van de Graaff accelerator) with intensities and other conditions such that they either simulate those in the reactor or provide a severe test of the stability under irradiation.

Test solutions in contact with Zircaloy-2 at temperatures in the range 60 to 120°C will be irradiated at power densities up to 150 w per cc. The solutions will be static in one type of experiment. In another type, the solutions will be circulated to produce film conditions comparable to those which will prevail in the reactor. The temperatures, power densities, solution compositions and container material are those of possible interest in the reactor. The use of electrons rather than reactor radiations is expected, from theoretical consideration, to increase the chance that an irradiation effect will occur at a given power density. The two different types of experiments are believed necessary to assure that the radiation stability is tested under conditions at least as severe as those in the reactor. Theoretical considerations did not enable us to predict reliably whether agitation would have any effect on the radiation stability or, in fact, the direction of an effect if one occurred.

The design for the static system is complete. Component testing to the extent feasible and desirable to provide reasonable assurance of design feasibility is also complete. Design and testing of the dynamic system has not been completed in like detail. However, we believe that the results of design work and of component testing which have been obtained provide sufficient basis for proceeding with the final design and construction.

Design information and results of component testing are summarized in this report.

2. Static System

2.1 General Description

The static system is comprised of an irradiation cell together with auxiliary equipment for the introduction of samples to be irradiated and for the recovery of the irradiated samples. A schematic drawing showing components and interconnections is given in Fig. 1.

The irradiation cell is comprised of a single loop of small bore Zircaloy-2 tubing surrounded by a coolant jacket, Fig. 2. The Zircaloy-2 tubing contains a fine metal filter at one end. The test solution will be exposed to radiation within the Zircaloy-2 tube. The dose rate may range up to 150 w/cc for exposure periods which may range up to many minutes. After exposure, the solution is forced through the filter, collected, and analyzed. The temperature during irradiation will be controlled by passing controlled-temperature water through the jacket at a rapid rate.

The auxiliary equipment consists of a solution reservoir, a solution displacement piston, and a sample collection chamber, together with connecting lines and valves. General descriptions of this equipment and of operating procedures were given in a previous report.[1] All equipment will be located within the Van de Graaff room.

More detailed information is presented below.

2.2 Specifications

2.2.1 Cell

A detailed drawing of the cell and of the mounting employed during irradiation is shown in Fig. 2. The dimensions of a given test system may differ slightly from those listed. In particular it now seems likely that the I.D. of the Zircaloy-2 test tube will be about 26 mils rather than 19 mils. These small differences in dimensions will not affect conclusions regarding the feasibility of the design.

Additional dimensions and parameters for the reference system of Fig. 2 are listed in Table 1.

2.2.2 Solution Reservoir

Materials and dimensions are given in Fig. 3.

2.2.3 Displacement Piston

The displacement piston is comprised of a titanium valve with Teflon packing manufactured by Autoclave Engineers, Erie, Pa. (30,000 psi series). Provisions will be made for remote operation using a step motor. A photograph of the motor-piston assembly is shown in Fig. 4. The displacement produced by turning the valve has been calibrated. Total displacement for seven complete valve stem turns was 0.10 cc. One complete valve stem turn required 200 steps on the motor control. This number of steps can be accomplished in as little as 0.7 min.

2.2.4 Sample Collection Chamber

A drawing with material and dimensions is shown in Fig. 5.

2.2.5 Valves and Connecting Lines

Valves which contact solution will be of titanium with Teflon packing

(Autoclave Engineers, 30,000 psi series) or stainless steel midget valves (Autoclave Engineers).

The tube connecting the cell with the collection chamber will be of 6 mil I.D., 304 stainless steel, 20 cm long. The remaining lines which contact solution will be of titanium or stainless steel with small bores and short lengths.

2.2.6 Coolant Heater

A drawing of the 1500 w heater which will be employed in controlling the coolant temperature is shown in Fig. 6. A manually operated Variac will probably be used to control the heating of the coolant during passage through the heater.

2.3 Procedures and Methods for Stability Determination

2.3.1 General Procedure

The planned procedure for accomplishing a stability experiment is as follows. A test solution of $CdSO_4$ is placed in the cell and irradiated at a specified power density and temperature for the desired time. The irradiated solution is then displaced into the sample collection chamber while irradiation continues. The sample is next recovered quantitatively from the chamber using up to 3 ml of wash solution to aid in the recovery. The sample is analyzed for Cd, and the stability during irradiation is evaluated from this analytical result together with prior information on the stability of the solution in the absence of radiation.

Detailed methods and procedures are described below.

2.3.2 Control and Determination of Solution Temperature

a. Introdution

The temperature of the solution is controlled by controlling the temperature of the outer surface of the Zircaloy-2 tube. This control of surface temperature is effected by passing controlled-temperature water through the coolant jacket at high velocity. Temperature gradients prevail in amounts which depend upon several factors, including (1) rates of heating in the different materials contained in the cell, (2) heat transfer properties including the calculated value of the film coefficient at the outer surface of the Zircaloy-2 tube, (3) velocity of coolant flow, and (4) temperature of the experiment.

An evaluation of temperature gradients is described below for the conditions: (1) 150 w/cc in solution, (2) 60°C and 120°C in solution, and (3) coolant velocity of 19 fps. A velocity of 19 fps at room temperature has been demonstrated with a mock-up in which the coolant jacket was attached to a laboratory water-tap supply. It is expected that somewhat higher velocities will prevail at higher temperatures.

b. Relative rates of heating in different cell materials.

The relative rates of radiation heating in the liquids, Zircaloy-2 and stainless steel which comprise the cell will depend upon the relative ranges and rates of scattering of electrons in the materials and upon the geometrical arrangement of the materials. For design considerations, it was assumed that the heating rate is inversely proportional to maximum range of electrons in the material. Closer estimates can probably be obtained from observations of the rise in temperature of the coolant during passage through the cell during an experiment. (The temperature of the coolant will be determined by means of thermocouples attached to the inlet and exit lines on the cell as indicated in Figure 2.)

The assumed values for relative heating rates at 150 w/cc in solution are the following.

Material	Heating rate (cal, sec^{-1}, cc^{-1})
Water	38
Zirconium (Zircaloy-2)	156
Stainless steel	226
Titanium	130.

c. Temperature gradients at 150 w per cc in solution

1. Heating rates in components of test loop

Calculated heating rates at 150 w/cc based on the above relative rates are set forth in Table 2.

2. Calculated temperature gradients

Calculated temperature gradients in the test loop at 60 and 120°C are listed in Table 3. One value for the temperature gradient across the film at 60°C was calculated for fully turbulent flow. Another was calculated for transition type flow. Since R_e is about 4400, it is considered likely that the coefficient will, in fact, correspond to that for transition flow. In any case, the calculated temperature drop across the film is small for flow of either type so that the uncertainty in the estimated solution temperature is not affected appreciably by this uncertainty. Thus, for example, if the calculated film coefficient is in error by a factor of two, the estimated solution temperature is in error by about 2.6°C at the maximum. This degree of uncertainty is considered acceptable.

For temperatures higher than 60°C, the flow will become more nearly turbulent, and the film coefficient will decrease as shown by the calculated value at 120°C.

For the portion of the tube containing the titanium filter it was assumed that no exchange of heat takes place between solution and filter and that the

Table 1. Reference Static Cell Dimensions

Component	Dimensions				
a. Zircaloy-2 tube for test solution	(cm)	(cc)	(cc/cm)	(cm^2/cm)	---
I.D.	.048	--	---	---	--
O.D.	.102	--	---	---	--
Internal volume per unit length (volume of solution)	--	--	.00183	---	--
Length of tubing irradiated	4.45	--	---	---	--
Total Internal volume	--	.00814	---	---	--
Internal area of tube per unit length	--	--	--	.151	--
Ratio of internal surface area to volume	--	--	--	---	83.5
Total volume of solution plus tubing and walls per unit length	--	--	.00816	---	--
Volume of wall per unit length	--	--	.00613	---	--
Area of outer wall per unit length	--	--	--	.320	--
b. Stainless steel cooling jacket					
I.D.	.137	--	--	---	--
Annular gap for H_2O	.018	--	--	---	--
Volume of H_2O per unit length of annulus	--	--	.00654	---	--
O.D.	.157	--	--	---	--
Total volume of tubing per unit length	--	--	.0193	---	--
Volume of wall per unit length	--	--	.0046	---	--
c. Titanium filter (density 3.80 g/cc)					
Thickness	.064	--	--	---	--
Diameter	.058	--	--	---	--
Pore size	.0003 to .00035	--	--	---	--

Table 2. Radiation Heating in Components of Reference Static Test Loop at 150 w/cc in Solution

Component	Heating Rate	
1. Test sample tube	$(cal,sec^{-1},cm^{-1})^a$	$(cal,sec^{-1},cm^{-2})^b$
Solution	0.07	0.22
Zircaloy-2 wall	0.96	2.99
Total of solution plus wall	1.03	3.21
2. Coolant jacket		
Water	0.25	--
Stainless steel	1.22	--
3. Total heat flowing into water (neglecting filter)	2.50	382[c]
4. Titanium filter	0.29[d]	--

a. Per unit length of coil.

b. Per unit area of outer surface of Zircaloy-2 tubing

c. Cal per sec per cc of water in annulus.

d. Assuming a density of 3.8 g/cc.

Table 3. Calculated Temperature Gradient in Static Test Loop
150 w/cc

Location	Temperature Gradient at 60°C	Temperature Gradient at 120°C[f]
1. Portion of Zircaloy-2 containing solution		
Center of solution to inner surface of tube	3.9[a]	3.9
Across Zircaloy-2 wall		
Volume heating in wall	1.6[b]	1.6
Heat from solution	0.2[b]	0.2
Total	1.8	1.8
Across film between Zircaloy-2 and coolant		
Turbulent flow	2.1 (h = 10,500)[c]	1.3 (h = 16,700)[g]
Transition flow	2.6 (h = 8,600)[c]	-[i]
Total from center of solution to coolant	7.8 to 8.3	7.0
2. Portion of tube containing titanium filter		
Center of titanium to inner surface of tube	0.7[d] (minimum)	0.7 (minimum)
Across Zircaloy-2 wall[e]	3.0 (maximum)	3.0 (maximum)
Across film between Zircaloy-2 and coolant		
Turbulent flow	2.1[c]	1.4[g]
Transition flow	2.8[c]	-[i]
Total from center of filter to coolant	5.8 to 6.5	5.1
3. Temperature rise in coolant at 19 fps	2.9	2.9

a. Appendix 1
b. Appendix 2
c. Appendis 3. The Re is 4400.
d. Appendix 4
e. Appendix 5
f. It was assumed that the values of all parameters were the same as those at 60°C except those influencing the film coefficient.
g. Re equal to 8,700. See Appendix 3 for method of calculation.
i. Flow is definitely turbulent.

heat from the filter flows through the ring of Zircaloy-2 in contact with the filter. In practice, exchange of heat between solution and filter will occur and the conduction area will exceed that in the ring. Accordingly, it is expected that the temperatures at given points in this region will probably be near those for similarly located points in other portions of the tube.

d. Summary

The temperature gradients and the degree of temperature control indicated by the above considerations are believed to be acceptable for the proposed experiments.

In practice, for the most precise estimates, temperature gradients must be calculated for each experiment using coolant flow rates and temperatures determined during an experiment, and also using best values for the dimensions of the particular experimental cell.

In the event that the degree of stability proves to be strongly dependent upon the temperature, it may be worthwhile to devise and perform experiments which will enable the solution temperature to be stated with greater precision.

2.3.3 Control and Determination of Power Density in Solution

a. Source of fast electrons

The ORNL Chemistry Division Van de Graaff accelerator will be employed in this work. A continuous, maximum, current of 100 μamps at 2 Mev can be obtained.

b. Electron beam intensity and distribution at cell

Measurements employing a multipoint collector[4] or a large collector area defined by a hole in a shield plate[1] showed that the intensity and distribution of the beam at a given total beam current is dependent upon the distance between the experimental cell and the Van de Graaff window and upon the material, size and position of beam scatterers placed between the cell and the Van de Graaff. The following arrangement has been selected for use with the static experiments.

Distance between cell and Van de Graaff window	2.27 cm

Scatterer centrally positioned

Material	Gold
Thickness	1 mil
Diameter	0.6 cm
Distance from cell surface	0.48 cm

Brass shield plate

Thickness	0.32 cm
Central hole diameter	1.27 cm
Separation between cell and outer Surface of plate	0.48 cm

The results[4] of tests of this arrangement using the multipoint collector without the shield plate indicated that the beam current will be substantially constant over the loop surfaces. They also indicated that the average current density will be about 30 $\mu amp/cm^2$ when the total current is 100 μamps.

c. Relationship between beam current and power density in solution

The results[5] of measurements in which a ceric dosimeter solution was used with a mock-up of the cell which conformed to the above arrangement led to the conclusion that the average power denisty in the ceric solution was 155 w/cc when the total beam current was 100 μamps. The estimated standard error in this value was $\pm$ 7 w/cc.

The planned experimental cell is similar to the mock-up, and it will be possible to estimate the power density at a given total current from the information obtained with the mock-up. In the event that the degree of stability of the solution proves to be strongly dependent on power density, it may be worthwhile to carry out additional experiments to improve the reliability and precision of power density values. Such experiments would employ a ceric solution within the experimental cell.

d. Recording of electron current

The current may vary to some extent during an exposure, and a record of the current will be obtained using a Sargent Recorder.

2.3.4 Methods and Sensitivity of Cadmium Analyses[6]

Small amounts of Cd such as those which may be obtained in static-type experiments will be analyzed polarographically. The expected accuracy is about ± 3% for concentrations of >3 μg/ml; at 1 μg/ml it is about ± 5%. Volumes of 5 ml are normally used, but a volume of 2 or 3 ml can be used without much additional effort. This accuracy is obtained in HCl solutions (0.1 M) but not in H_2SO_4 solutions.

To place this information in perspective, assume that we have a 0.008 ml sample of a 0.01 M $CdSO_4$ solution collected in the sample collector. The sample then contains 8.8 μg of Cd. This sample is removed from the collector using 0.1 M HCl to wash the collector surfaces and thus aid in obtaining complete recovery. The sample is diluted to a total volume of about 3 ml using 0.1 M HCl, and then analyzed polarographically to an overall accuracy of about ± 0.3 μg in the 8.8 μg sample. Since the area of Zircaloy-2 in contact with the sample is 0.67 cm^2, the sensitivity of the determination of the amount adsorbed is ± (1.4) (0.3)/0.67 = ± 0.65 $\mu g/cm^2$.*

It is interesting to note that the sensitivity of the determination of the amout adsorbed would not be increased and may be decreased by obtaining a larger sample. Thus, if the sample contains 100 μg, the sample would be analyzed coulometrically with an expected sensitivity of about ± 1 μg so that uncertainty in the amount adsorbed would be about ± (1.4)(1.5) $\mu g/cm^2$ = ± 2.1 $\mu g/cm^2$.

2.3.5 Control Tests of Solution Stability

Various control experiments will be required to establish the stability of the $CdSO_4$ test solution in the absence of radiation. The experiments which are now planned are listed below along with explanatory remarks.

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* The factor 1.4 enters because the final result is the difference between two values known with equal accuracy

a. Determination of the amount of Cd adsorbed on surface of 6 mil stainless steel capillary in contact with the particular solution to be used in test.

This determination is needed to establish whether it is possible in principle for desorption of Cd from the tube which connects the cell and sample collector to significantly change the concentration of Cd in the irradiated solution as it passes through the tube.

It is expected that the amount adsorbed will be $< 0.5\ \mu g/cm^2$ or a total of $< 0.5\ \mu g$ on the $0.9\ cm^2$ of tube surface. Since the expected sensitivity of Cd detection in the sample will be about $\pm 0.3\ \mu g$, it can be assumed that desorption from the stainless steel will not affect the test results when the amount on the steel initially is $0.5\ \mu g/cm^2$ or less. In practice, we are not certain that the sorption will be $< 0.5\ \mu g/cm^2$ in a given solution, and we need to verify this.

Sorption on the stainless steel tubing from the sample would affect the results if it occurred to an appreciable extent. However, it is reasonable to assume that the steel surfaces will have been nearly saturated with Cd prior to the test so that little or no additional absorption will occur during the time the test solution is in contact with the steel.

The planned procedure is as follows:

Pass $CdSO_4$ test solution through a length of stainless steel tubing for a short time and then allow the solution to stand, at room temperature, for about 30 min.

Force solution out of tube and rinse tube with distilled water.

Rinse tube with 0.01 M HNO_3 and submit rinse solution for analyses.

b. Determination of amount of Cd adsorbed on Zircaloy-2 test tube at room temperature.

The planned procedure for this test is the same as that described above.

c. Determination of concentration of Cd in solution placed in cell.

The planned procedure is as follows:

Assemble irradiation cell and auxiliary equipment. Charge with solution. Collect sample in collector. Remove sample using 0.1 M HCl wash solution to aid in recovery of sample in collector. Analyze sample. Repeat collection and analyses of samples until the observed concentration remains constant.

d. Determination of amount of Cd adsorbed on Zircaloy-2 test tube at test temperature.

The planned procedure is as follows:

Use the assembled irradiation cell and auxiliary equipment. Charge with solution sample. Heat cell to test temperature and allow to stand for 30 min. Collect sample and analyze. Repeat procedure until observed concentration remains constant.

2.4 Status of Equipment

A mock-up of the cell without filter has been constructed and used in dosimetry experiments.[5,7] Experience with this mock-up showed that the cell can be constructed according to design. A technique has been developed by which titanium filter material, 3.5 micron mean pore size and 0.030 in. thick, can be press-fitted into the Zircaloy-2 tubing at the proper location. Microscopic examination after grinding away a section of the tube and filter indicated that the filter was intact. Additional examinations will be made to estimate the tightness of the seal between the filter and the tube walls.

At present only the titanium filter material is on hand. However, porous zirconium material of 20-30 micron mean pore size and $\sim$ 0.090 in. thick has been ordered. This is the only porous zirconium available and although the mean pore size is somewhat large it may prove useful in some experiments.

3. Dynamic System

3.1 Introduction

Dynamic experiments will be conducted with a small, high speed (35,000 rpm), centrifugal pump with which solution is circulated through a small bore tube which forms a loop in front of the cover plate of the pump. The diameters of the pump cavity and of the tube bore are about one-half in. and 26 mils. The total fluid volume is about one-fourth cc and all of the fluid will be irradiated continuously during an exposure.

The purpose of the tube is to provide a channel in which film conditions can be made comparable to those in the reactor. The flow around the impeller and housing cannot be well defined, and the film conditions in this region cannot be estimated reliably. The selection of the bore diameter was based on: (1) calculated values for the solution velocity required in a tube of given bore diameter in order to establish film coefficients comparable to those in the reactor at a velocity and hydraulic diameter of 40 fps and 100 mils, (2) experimental values for head-flow characteristics of a pump which has been designed and tested, and (3) experimental values for the flow rate-pressure drop relationship in a 26 mil I.D. tube bent into the shape required with the cell.

The bases for limiting the pump diameter to one-half in. and for the other pump design features included: (1) the assumption that the dose rate should be approximately uniform through the solution, (2) experimental information which showed that suitable doses and approximately uniform dose rates can be achieved with the proposed design and with the available Van de Graaff,[5] and (3) temperature control considerations.

The method of performing an experiment with this system is similar to that previously described for the static system. After the solution is irradiated for a desired period, it will be diluted and forced through a filter using fresh solution. Irradiation and circulation will continue during this dilution. After an amount of solution sufficient for accurate analyses (see Sec. 3.7) has

been forced from the cell, the irradiation and circulation are stopped. Loss of Cd during irradiation is determined from the results of analyses of one or more of the following: (1) solution drawn from the cell during operation, (2) solution remaining in the cell at termination of irradiation, and (3) Cd remaining on cell surfaces after the solution is withdrawn. In the event the solution is unstable during irradiation and the instability reverses rapidly after termination of irradiation, we will not be able to determine whether the instability occurs in the tube or in the pump or in both regions. However, it is expected that the primary objective will be achieved; that is, a determination of whether the solution is stable under dynamic conditions which are at least as severe as those which will prevail in the reactor.

The auxiliary equipment consists of the displacement piston, solution reservoir, and sample collection chamber which were described in connection with the static system. The interconnections are also basically the same as those described for the static system. However, all of this equipment except the sample collection chamber will be located outside the Van de Graaff room. The coolant heater will also be used with the dynamic system.

More detailed information on design, component testing, and procedures is presented below.

3.2 Pump Performance

A pump of the dimensions and speed needed or permissible with the dynamic system has been designed, and tests of performance have been made using a stainless steel model.* Figure 7 is a drawing of the pump and Fig. 8 is a photograph of the partially assembled pump showing the impeller, seal, housing and drive motor. Head-flow characteristics were measured by operating the pump in a test stand with equipment arranged as shown in Fig. 9. Inlet and outlet lines of

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* The test model was designed by L. V. Wilson of ORNL Reactor Division.

0.040 in. I.D. capillary tubing, approximately 4.5 cm total length, were used in the test. These tubes were located between the pump and the pressure gauges.

The results of measurements of the drop in pressure across valve No. 3 over a range of volume flow rates are shown by lines A, B, and B' in Fig. 9. The changes in flow rates for the line A data were effected by adjustment of valve No. 3 while the pump speed was held constant at 33,000 rpm. The changes in flow for the data of lines B and B' were produced by varying the pump speed with constant valve settings. The head-flow characteristics of the pump which were estimated from these data and from the results of calculations of the pressure drop in the small tubing and connections are shown by line C. The two points through which this line was drawn are also shown. At 33,000 rpm the shut-off head was about 77 ft. The head decreased with increasing flow and was about 51 ft at 220 cc/min.

3.3 Loop Dimensions

Shown in Fig. 10 are calculated values for the solution velocity required in a tube of given diameter in order to establish film coefficients comparable to those in the reactor at a velocity and hydraulic diameter of 40 fps and 100 mils. The values of the upper curve were calculated for straight tubes, (Appendix 5). Those for the lower curve were calculated[3] for a curved tube of 0.6 cm radius. As a result of these calculations, it is expected that a velocity of about 23 fps will be sufficient for the purpose of the experiment.

The results of pressure drop-volume flow measurements in a 26 mil I.D. tube bent into the shape required with the cell are shown in Fig. 11.* The available heads at different flow volumes from Fig. 10 are also shown in Fig. 11. The results show that a velocity of about 31 fps would be produced by the present

*The discharge of the tube was directly at atmosphere or to atmosphere through a 1 in. length of 80 mil I.D. plastic tubing.

pump design in a loop constructed of 26 mil tubing.

Although it appears likely that the minimum required velocity of 23 to 24 fps could be obtained with the available pump in tubes having bores greater than 26 mils, no appreciable advantage would result from the use of larger bores. Accordingly, we plan to use a bore diameter of about 26 mils.

3.4 Summary of Cell Dimensions

A drawing of the dynamic cell showing important dimensions is given in Fig. 12. Additional cell parameters are listed in Table 4.

3.5 Power Density

Measurements of electron currents from the Van de Graaff using a multi-point collector (exp. 8 of ref. 4) showed an arrangement of cell and electron scatterer for which the current density is approximately uniform over the surface of a 1.3 cm disk. Subsequent measurements[5] using a ceric dosimeter solution in a system which simulated the materials and dimensions of the dynamic system showed that a power density of 150 w/cc can be obtained with the cell and scatterer positions referred to above. The final cell design will differ sufficiently from the mock-up that it will be worthwhile to perform additional dosimetry experiments in which the ceric solution is exposed within the experimental cell when the cell becomes available. The power density prevailing in a given experiment will be determined from the power density-beam current relationship found in these additional measurements.

It may be noted that the power density in the solution within the tube may differ from that in the remainder of the solution owing to differences between the amount of absorber between the Van de Graaff and the solution and to differences between the amounts of scattering of electrons from Zircaloy-2 walls into the solutions. It can be speculated that the difference in power density will be

Table 4

Some Dynamic Cell Parameters

Tubing Loop	
Length	3.8 cm
Bore diameter	.066 cm
Volume	
per unit length	.0034 cc/cm
total	.013 cc
Internal surface area	
per unit length	.21 cm^2/cm
total	.80 cm^2
Wall thickness	2 to 3 mils
Reynolds number at 24 fps and 60°C	10,000
Pump Cavity	
Diameter	1.27 cm
Surface area (approximate)	
front and back surfaces	3.45 cm^2
outer edge of cavity	0.41 cm^2
vanes	0.64 cm^2
total	4.5 cm^2
Volume (approximate)	0.25 cc
Power in cell at 150 w/cc of solution which must be removed by coolant (approximate)	85 w

small. However, a measurement of the power density within the solution in the tube will also be made.

The total power deposited within the cell is expected to be about 85 w at 150 w per cc in solution. This estimate of power generation is based on the average current density which was determined from exp 8 of ref. 4.

3.6 Temperature Control

Estimates have been made (Appendix 6) of heat which can be removed from the circumference of the impeller housing. It was assumed that heat would pass through 5 mils of Zircaloy-2 at the circumference and into a contacting aluminum jacket. The temperature of the jacket would be controlled by circulation of a coolant. It was found that all of the radiation heat in the cell can be removed in this way when (1) the cell temperature is 60°C, (2) the velocity in the loop is 24 fps, and (3) the coolant temperature is about 20°C. For higher cell temperatures and/or higher velocities in the loop, the coolant temperature can be higher than 20°C. Accordingly, it was concluded that the temperature of the cell can be controlled by passing controlled temperature water through the aluminum jacket.

The previously described heater will be employed in controlling the temperature of the coolant.

Estimation of the temperature gradients in the circulating solution show that at 150 w per cc and at 31 fps in the tube, the temperature rise in solution passing through the pump is about 6°C. The rise for solution passing through the loop is about 0.4°C if no heat is lost to surrounding air. Temperature gradients at other operating conditions can be estimated readily from these values.

3.7 Methods and Sensitivity of Cd Analyses[6]

As previously mentioned, the volume of the dynamic system will be about 0.25 ml, and the area of the 26 mil tube will be about 0.8 cm^2. A solution sample will be obtained during irradiation by dilution of the solution within the cell. The solution within the cell can also be removed for analyses after irradiation.

The samples will be analyzed using controlled potential coulometry. The expected accuracy is $\pm$ 1 µg at the 100 µg level, and $\pm$ 0.3% at 300 to 1000 µg level.

To put these values into perspective, assume that the solution within the cell contains 275 µg of Cd (0.25 ml of 0.01 M $CdSO_4$) and that 300 µg are contained in the solution which has passed through the cell and which is employed in the analyses. The uncertainty in the total amount of Cd in solution will then be $\pm$ (575)(.003) = $\pm$ 1.7 µg.

Assuming that Cd will be adsorbed only on the 0.8 cm^2 of tube surface the uncertainty in the amount adsorbed will be $\pm$ 1.7 / 0.8 = $\pm$ 2.1 µg/cm^2.

It may be noted that the sensitivity of the determination of adsorption increases somewhat as the concentration of Cd in the sample decreases.

It may also be noted that the sensitivity in the above example can be increased by determining the Cd retained in the cell after an exposure. Thus, assume that adsorbed Cd returns to solution after termination of irradiation. This redissolved Cd is retained within the .25 ml of solution within the cell. Assuming further that the amount of Cd in the original solution is 275 µg per 0.25 ml and that the cell solution is brought to the original concentration by passing a sufficient volume of solution through the cell, the uncertainty in the amount redissolved is $\pm$ (1.4)(275)(.003) = $\pm$ 1.2 µg or $\pm$ 1.4 µg per cm^2 of tube surface.

If the adsorbed Cd does not redissolve readily in the solution after irradiation, the amount adsorbed can be determined with high accuracy. Thus assume that deposited Cd is dissolved in 3 cc of acid solution containing no other Cd. The expected sensitivity of analyses of the small amount of Cd is about $\pm$ 0.15 μg/ml so that the total uncertainty in the amound adsorbed would be $\pm$ 0.45 μg($\pm$ 0.56 μg/cm^2 on 0.8 cm^2).

3.8 Control Experiments

Control experiments will be performed to determine adsorption in the absence of radiation. Essentially the same procedures as those planned for the radiation experiments will be used.

4. Test Schedule.

A provisional testing schedule for the first experiments to be carried out is given in Table 5.

Table 5

Provisional Testing Schedule

Test No.	Exposure		Solution Composition			Temperature
	Dose Rate (w/cc)	Time (min)	[$CdSO_4$] (m)	pH with H_2SO_4	Gas Content	(°C)
1	150	10	0.04	(a)	He	60
2	150	30	0.04	(a)	He	60
3	150	30	0.04	(a)	He	120
4	150	10	0.01	(a)	He	120
5	150	30	0.01	(a)	He	120
6	150	10	0.01	(a)	He	60
7	150	30	0.01	2	He	60
8	150	30	0.01	2	He	60
9	150	30	0.01	2	He	60

(a) No acid added.

The primary objective of this series of tests is to orient ourselves with regard to the major variables likely to influence the solution stability. Depending on the directions indicated by the results obtained, we will then make a more detailed study of the important parameters over the ranges of interest; i.e.,

Concentration	$CdSO_4$	0.01 - 0.1 M
pH		2 - 5
Temperature		60 - 120°C
Dose rate		to 150 w/cc
Exposure time		1 to 30 min.
Hydrogen content		0 to 15 psi, equivalent

Experiments with the dynamic system will depend upon the results obtained in static tests; although with suitable duplication to evaluate flow effects.

Appendix 1

(Static System)

CALCULATION OF ΔT ACROSS SOLUTION IN ZIRCALOY-2 TUBE AT 150 w/cc.

Use equation,

$$\Delta T = \frac{qr^2}{4\ k}$$

q = rate of volume heating in solution

r = internal radius of Zircaloy-2 tube

k = thermal conductivity of solution

Introducing values

$$\Delta T = \frac{(38)(0.024)^2}{(4)\ (1.4 \times 10^{-3})}$$

$$\Delta T = 3.9°C$$

Appendix 2

(Static System)

CALCULATION OF TEMPERATURE DROP ACROSS ZIRCALOY-2 WALLS AT 150 w/cc.

Average area for conduction in Zircaloy-2 wall

$$\bar{A} = \frac{A_2 - A_1}{\ell n\ A_2/A_1}$$

Introducing values

$$\bar{A}/cm = \frac{0.320 - .151}{\ell n\ \frac{.320}{.151}}$$

$$\bar{A}/cm = 0.225\ cm^2/cm$$

Rate of radiation heating in wall per unit average area

$$q = \frac{cal,sec^{-1},cm^{-1}}{(cm^2,cm^{-1})}$$

$$q = \frac{0.96}{0.225}$$

$$q = 4.3\ cal,sec^{-1},cm^{-2}$$

Temperature gradient across wall resulting from heating in walls

Use equation,

$$\Delta T = \frac{qL}{2k},$$

where L is the thickness and k is the thermal conductivity of Zircaloy-2

Introducing values

$$\Delta T = \frac{(4.3)(.027)}{(2)(.036)}$$

$$\Delta T = 1.6°C$$

Temperature gradient across wall resulting from heating in solution

$$\Delta T = \frac{qL}{k\bar{A}}$$

where q is the heat from solution (cal, sec^{-1}, cm^{-1}), and L is the wall thickness.

Introducing values

$$\Delta T = \frac{(.07)(.027)}{(.036)(.225)}$$

$$\Delta T = 0.23°C$$

Appendix 3

(Static System)

CALCULATION OF FILM COEFFICIENT IN COOLANT JACKET AT 19 fps and 60°C.

1. Dimensionless numbers

   a. Reynolds number

$$Re = \frac{DV}{\gamma}$$

$$Re = \frac{(.036)(580)}{.0047}$$

$$Re = 4,400$$

b. Prandtl number

$Pr = 3.02$

Other quantities

a. Specific weight, γ, $= 61.52$ lb/ft^3

b. Specific heat, $Cp = 1.00 \frac{Btu}{(lb)(F)}$

2. Evaluation of film coefficient for well established turbulent flow

(1)[a] $\frac{h}{\gamma\ CpV} = \frac{Nu}{RePr} = \frac{0.0396(Re)^{-1/4}}{1+A(Re)^{-1/8}(Pr-1)}$

Where $A = 1.7$

Evaluating the right side of the equation, we have,

$$\frac{h}{\gamma\ CpV} = 2.21 \times 10^{-3}$$

but $\gamma CpV = 4.2 \times 10^6 \frac{Btu}{(hr)(ft^2)(F)}$

then $h = 9{,}300 \frac{Btu}{(hr)(ft^2)(F)}$

Multiplying this value by the factor,[b]

$1 + 1.77\ d/R,$

which accounts for the effect of curvature, we have

$$h = 10{,}500 \frac{Btu}{(hr)(ft^2)(F)}$$

3. Evaluation of film coefficient for transition zone flow.

In this case we again use Eq. 1 above but the Nusselt number is that given by the Hausen expression

(2)[b] $Nu = 0.116(Re^{2/3}-125)Pr^{1/3}[1 + (\frac{d}{L})^{2/3}](\frac{u_B}{u_W})0.14$

where u_B and u_W are the viscosities of the water at the temperatures of the bulk solution and of the wall.

Evaluating this expression we have,

$$Nu = 24.2$$

then from Eq. 1

$$\frac{h}{\gamma\ CpV} = 1.82 \times 10^{-3}$$

then h in a straight channel is

$$h = 7{,}600.$$

Increasing this value by the curvature factor, we have

$$h = 8{,}600\ \frac{Btu}{(hr)(ft^2)(F)}$$

a. Ref. 2
b. Ref. 3

Appendix 4

(Static System)

CALCULATION OF ΔT FROM CENTER OF TITANIUM FILTER TO ZIRCALOY-2 WALL

Assume that heat is not exchanged with solution

$$\text{then}\quad \Delta T = \frac{qr^2}{4k}$$

Introducing values

$$\Delta T = \frac{(130)(.029)^2}{(4)(0.04)}$$

$$\Delta T = 0.7°C$$

Appendix 5

(Static System)

CALCULATION OF TEMPERATURE DROP ACROSS ZIRCALOY-2 WALLS AT POINTS OF CONTACT WITH TITANIUM FILTER AT 150 w/cc.

Assume that heat from filter flows only in a radial direction

Also assume that the average area for heat conduction is the same as that employed for other parts of the tube (Appendix 2) although the wall thickness here is only 0.022 cm.

Temperature drop across wall from volume heating in wall is then assumed to be 2.2°C.

Calculate ΔT resulting from heat flowing from filter

$$\Delta T = \frac{qL}{k\bar{A}}$$

Introducing values

$$\Delta T = \frac{(0.29)(.022)}{(.036)(.225)}$$

$$\Delta T = 0.8°C$$

Appendix 6

(Dynamic System)

VELOCITY REQUIRED IN STRAIGHT TUBES OF 25 to 40 mil BORE IN ORDER TO SIMULATE FILM CONDITIONS AT VELOCITY OF 40 fps AND A HYDRAULIC DIAMETER OF 100 mil.

According to Kent's Handbook[a] the film coefficients in tubes of different hydraulic diameters are related to the diameters and velocities by the equation,

$$h = k \frac{V^{0.8}}{d^{0.2}},$$

where k is a constant for a given fluid and temperature, V is the velocity, and d is the hydraulic diameter. This relationship was employed in calculating the values for the upper line in Fig. 11.

[a]Ref. 8

Appendix 7

(Dynamic System)

ESTIMATE OF AMOUNT OF HEAT WHICH CAN BE REMOVED FROM CIRCUMFERENCE OF IMPELLER HOUSING

Estimates of the film coefficient for this surface were based on the assumptions: (1) that the linear velocity of solution in the annular region adjacent to the inner circumference will match the peripherial velocity of the impeller, and (2) that the film conditions near the circumference will be comparable to those for flow in a straight pipe at the same velocity. A list of parameters employed in the estimates of heat removal follows.

Values for the film coefficients calculated using Eq. 1 of Appendix 3 are included in this tabulation.

	Value of parameter	
Parameter	24 fps	31 fps
Impeller diameter	.454 in.	.454 in.
Hydraulic diameter of annular region	.033 in.	.033 in.
Pump speed	25,000 rpm	33,000 rpm
Peripherial velocity of impeller	49 ft per sec	65 ft per sec
Reynold's number at above diameter and velocity and at 60°C	26,000	35,000
Heat transfer area	4.36×10^{-4} ft^2	4.36×10^{-4} ft^2
Thickness of Zircaloy-2 between solution and aluminum jacket	.005 in.	.005 in.
Film coefficient (Btu/hr·ft^2·°F)	18,000	23,000

Now for housing wall thickness of 0.005 in.,

$$h = K/X = \frac{8.3}{.005}\ 12 = 20{,}000 \text{ Btu/hr·ft}^2\text{·°F}$$

At 31 fps in tube

$$\frac{1}{U} = \frac{1}{23{,}000} + \frac{1}{20{,}000} = 9.4 \times 10^{-5}$$

$$U = 11{,}000 \text{ Btu/hr·ft}^2\text{·°F}$$

Heat removed from housing

$$UA\Delta T = 11{,}000 \times 4.36 \times 10^{-4} \times \Delta T = 3.8\ \Delta T \text{ Btu/hr}$$

At 24 fps in tube

$$\frac{1}{U} = \frac{1}{18{,}000} + \frac{1}{20{,}000} = 1.06 \times 10^{-4}$$

$$U = 9{,}500 \text{ Btu/hr}\cdot\text{ft}^2\cdot{}^\circ\text{F}$$

Heat removed from housing

$$UA\Delta T = 9{,}500 \times 4.36 \times 10^{-4} \times \Delta T = 4.1\ \Delta T \text{ Btu/hr}$$

For 85 watts heat input (290 Btu/hr)

$$\Delta T = \frac{290}{4.8} = 60^\circ\text{F at 31 fps in tube}$$

$$\Delta T = \frac{290}{4.1} = 71^\circ\text{F at 24 fps in tube}$$

Considering the results of these estimates and of the facts that undetermined amounts of heat will be lost from the face of the pump and from the shaft, it was concluded that the temperature can be controlled by transfer of heat through the pump housing to a water cooled aluminum jacket.

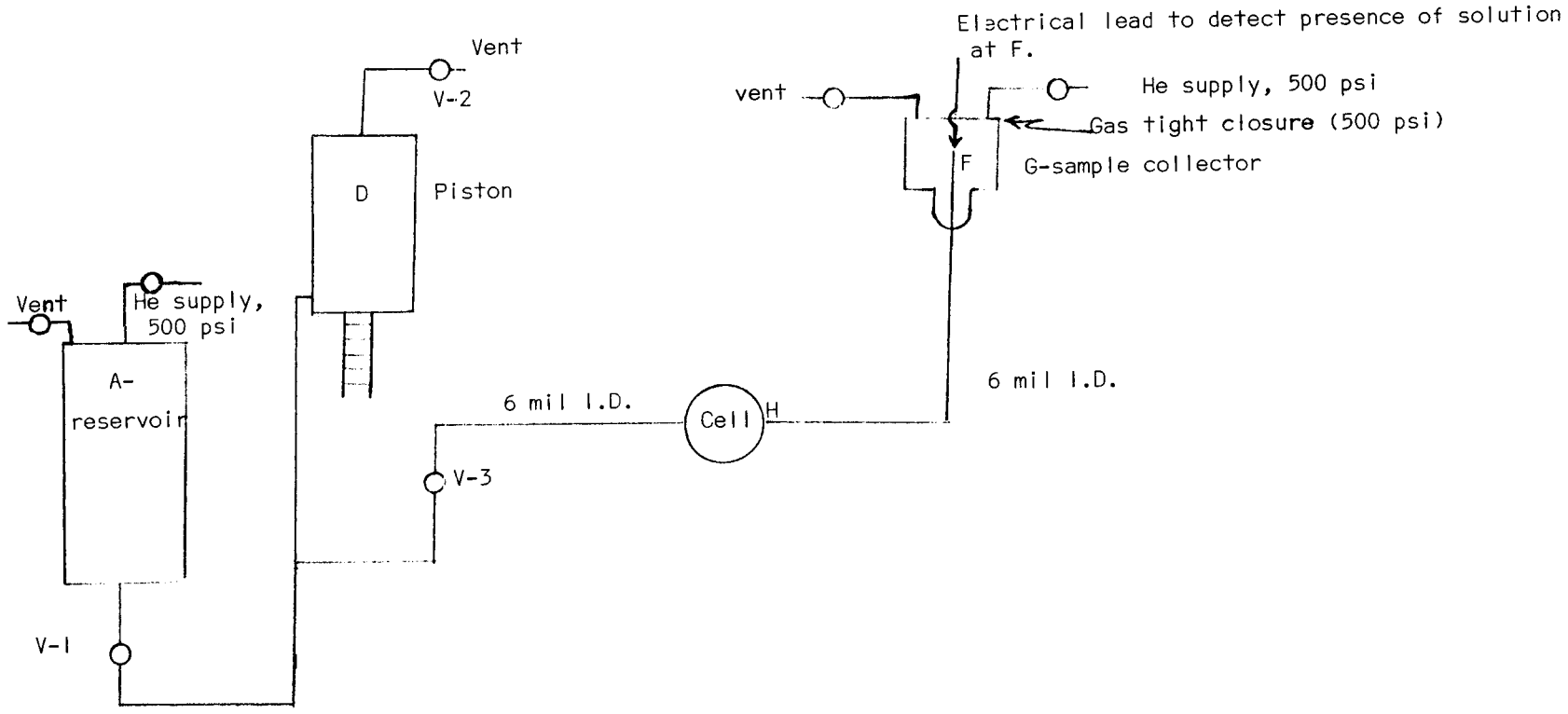

Fig. 1. Equipment for Irradiating Small Volumes of Solution in Static System.

Part No.	Part Name	Material	Dimensions
1	Test tube	Zircaloy-2	19 mil I.D.; 40 mil O.D. tubing Coil diameter of 1/2-in. Lenght of irradiated portion, 1-3/4-in. Test tube is mounted such that no thermal contact exists with mounting or shield plates.
2	Cooling jacket	Stainless steel	62 mil O.D.; 54 mil I.D. tubing.
3	Solution inlet-outlet	Stainless steel	6 mil I.D.; 50 mil O.D. Sealed to test tube mechanically.
4	Shield plate	Brass	3-in. diam. x 1/8-in thick spaced so that it clears test coil. Hole is 1/2-in.diam.
5	Controlled temperature water adapter	Stainless steel	62 mil O.D.; 40 mil I.D. tubing.
6	Thermal Insulator	Micarta	1/16-in. thick placed to prevent thermal contact between test coil and mounting plates.
7	Clamp	Brass	1-3/4-in. x 3/4-in. x 1/8-in.
8	Clamp	Brass	5/8-in x 5/8-in. x 1/4-in.
9	Water cooling tube	Copper	3/16-in. O.D.
10	Electron beam scatterer	Gold	1 mil thick, 1/4-in. diam. located 3/16-in from plane of coil.
11	Water cooling tube	Copper	3/16-in. O.D.
12	Mounting Plate	Brass	4-1/2in. diam x 1/4-in. thick.
13	Adapter	Stainless steel	Joints sealed with epoxy cement.
14	Filter	Titanium	25 mil thick.

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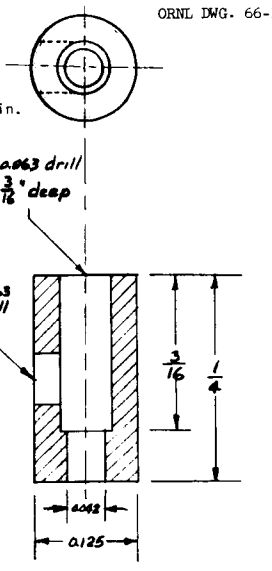

DETAIL ITEM (13)

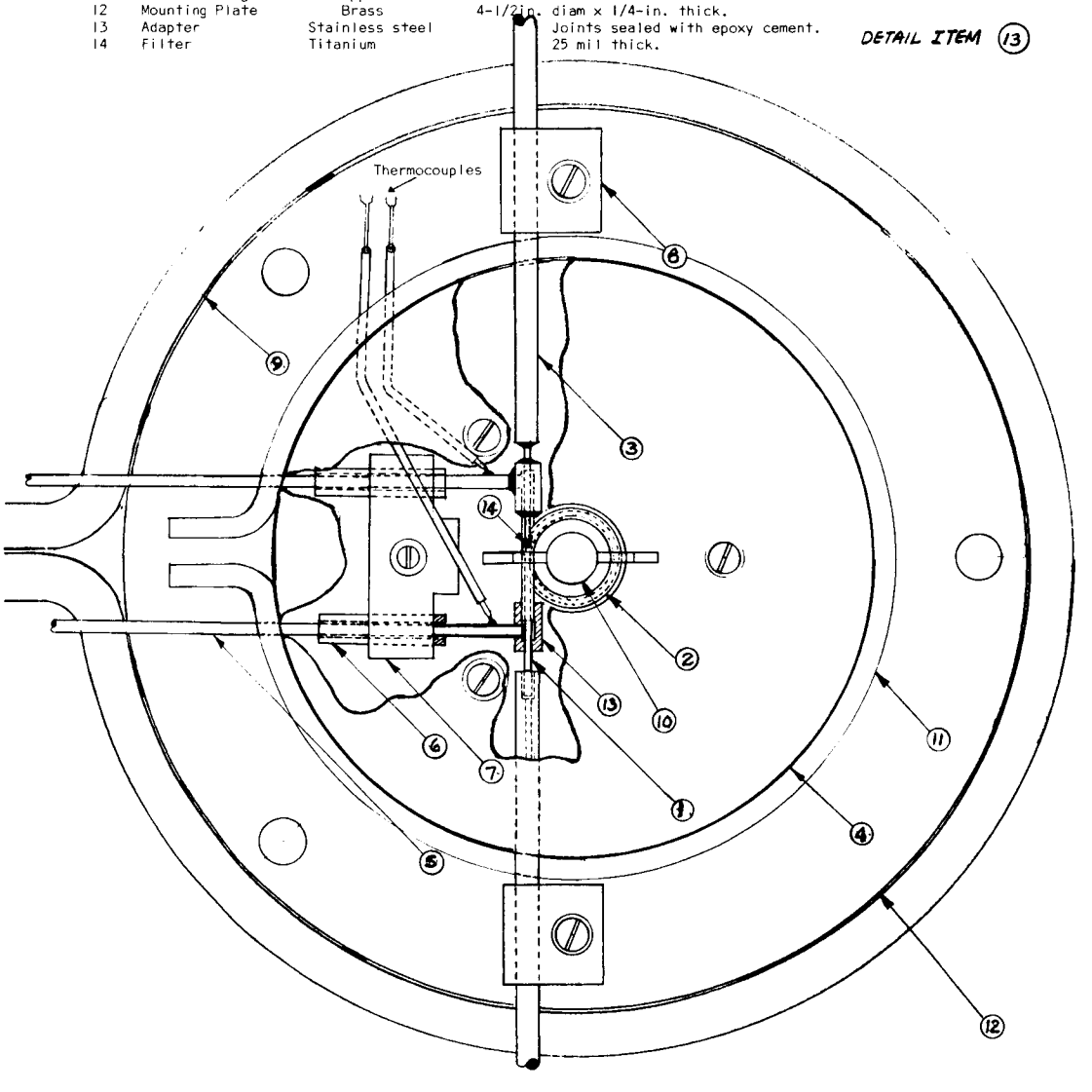

Fig. 2. Static Irradation Cell.

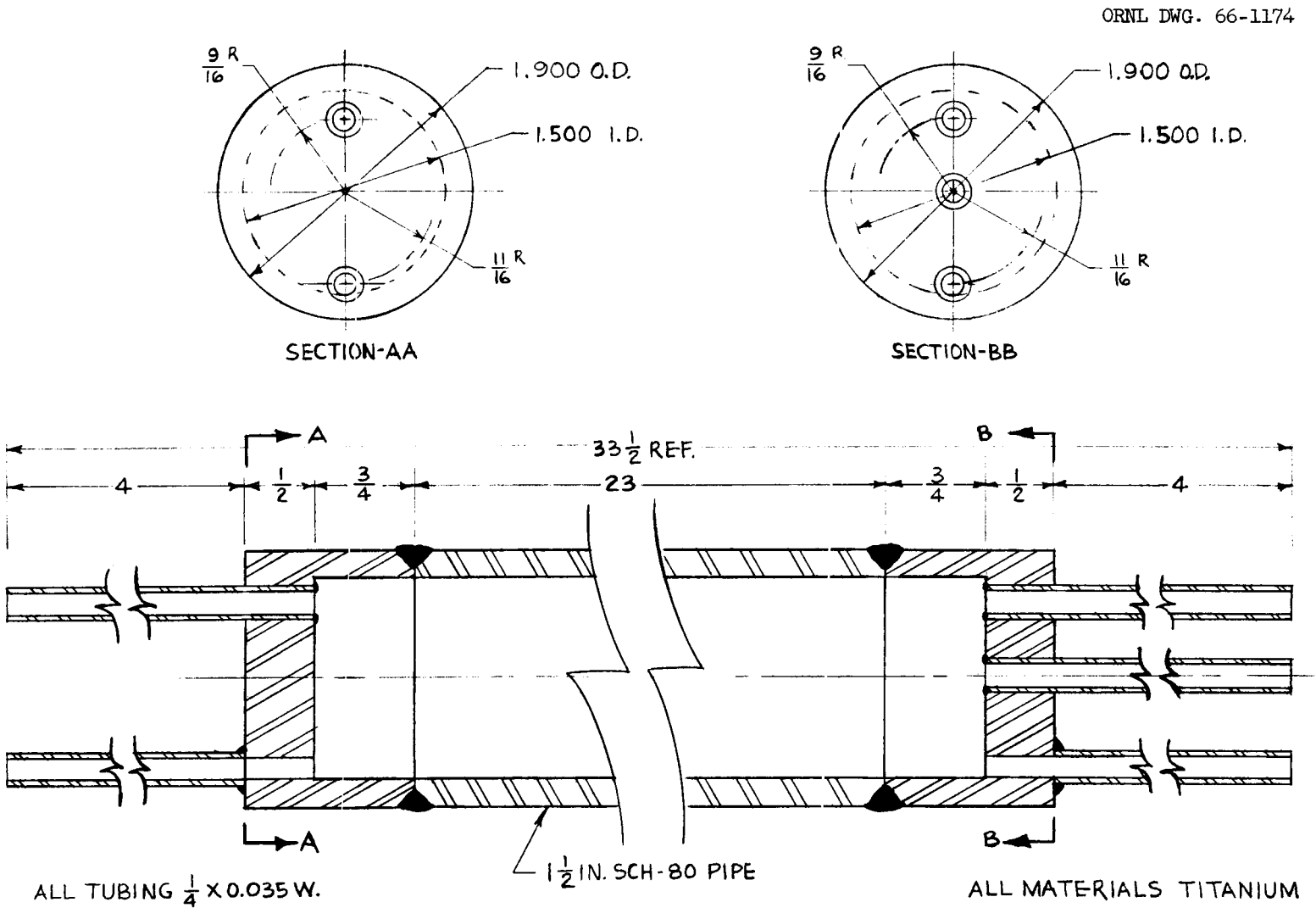

Fig. 3. Solution Reservoir.

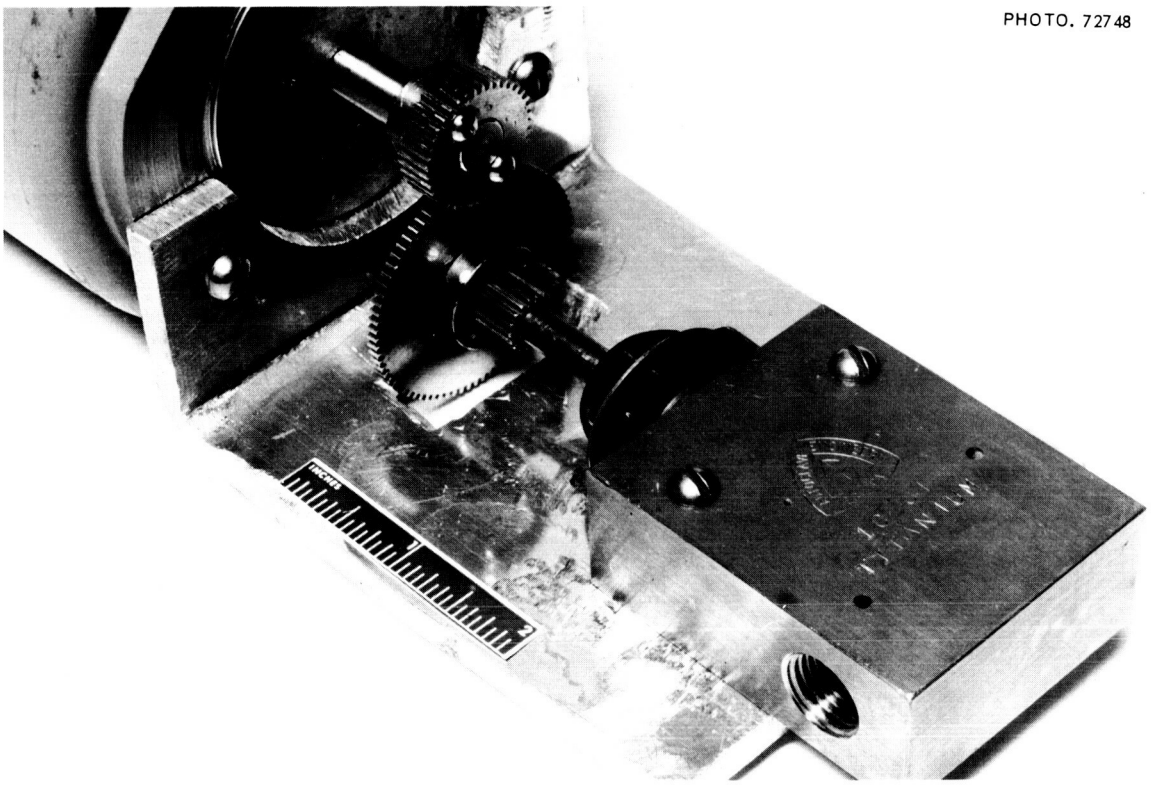

Fig. 4. Photograph of Titanium Valve (Displacement Piston), Step Motor and Drive Gears.

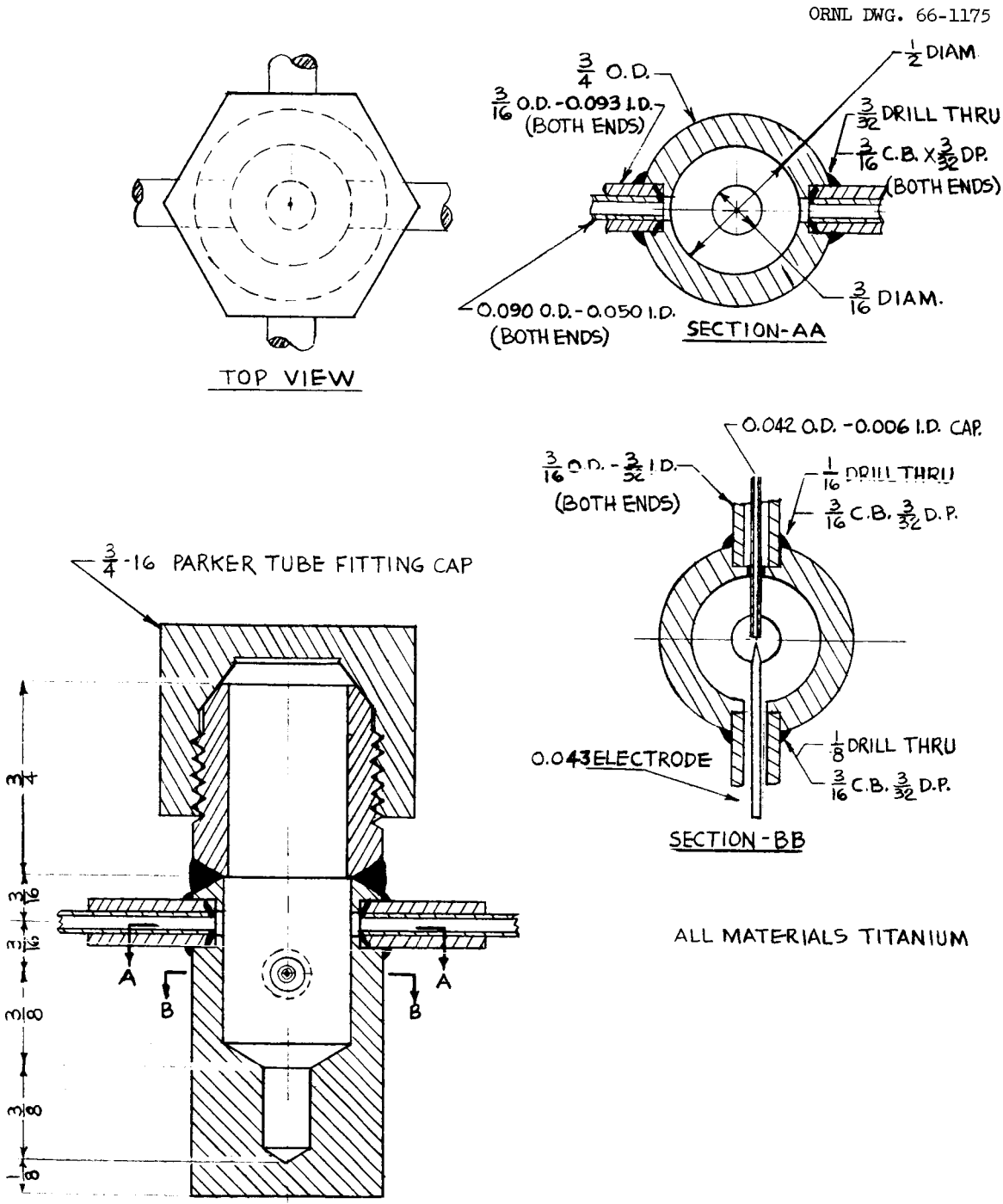

Fig. 5. Sample Collection Chamber.

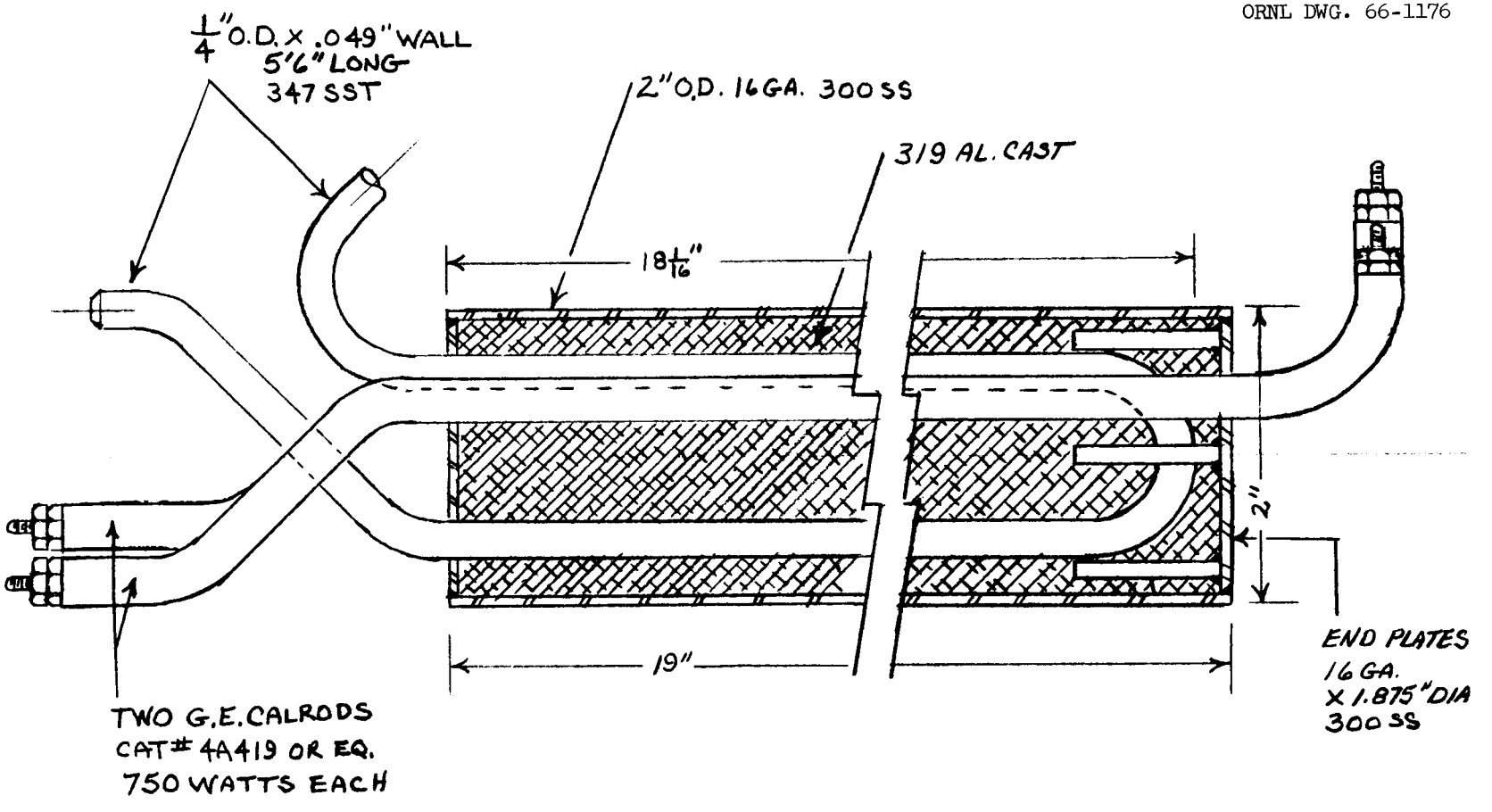

Fig. 6. Coolant Temperature Controller.

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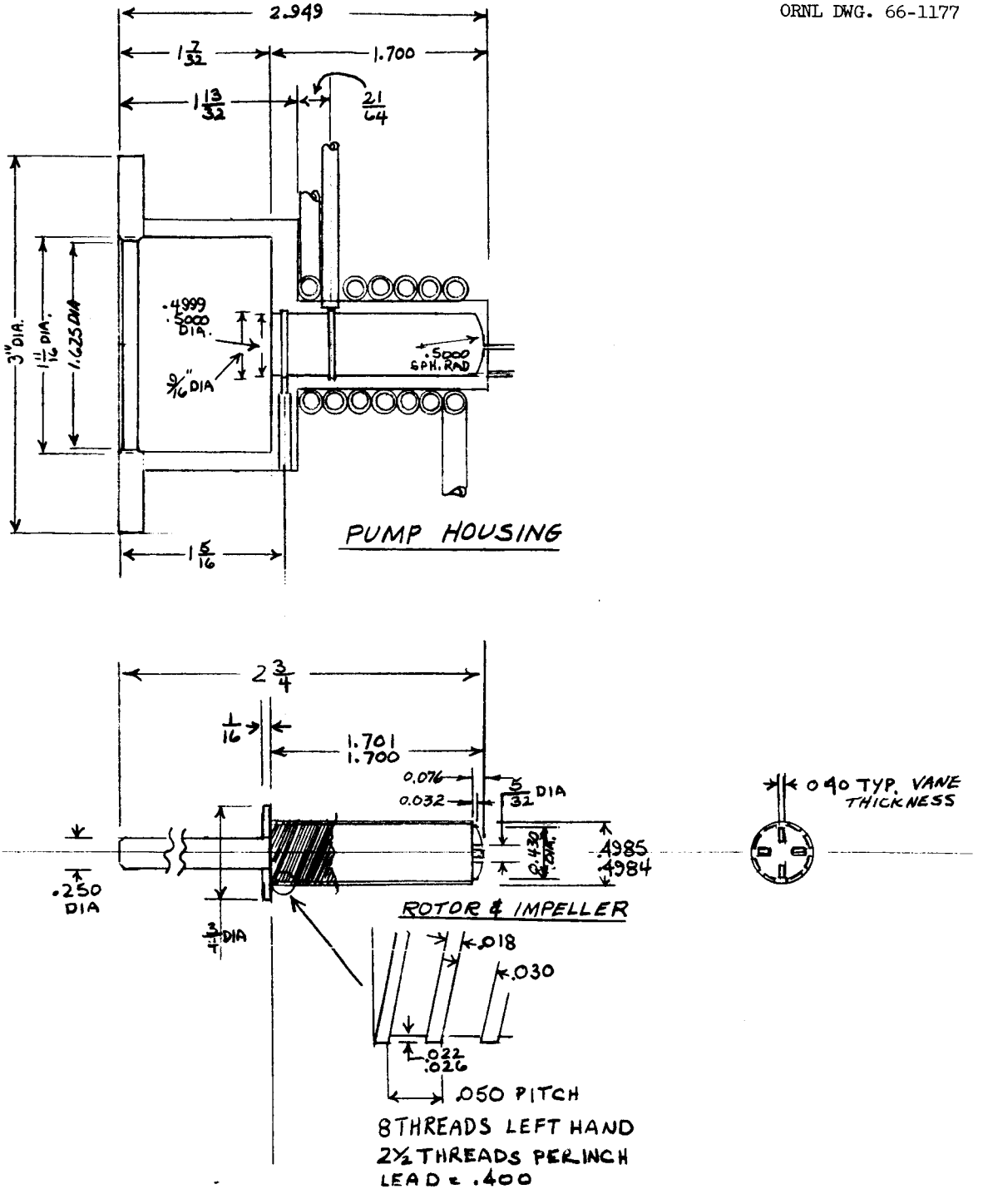

Fig. 7. Test Pump.

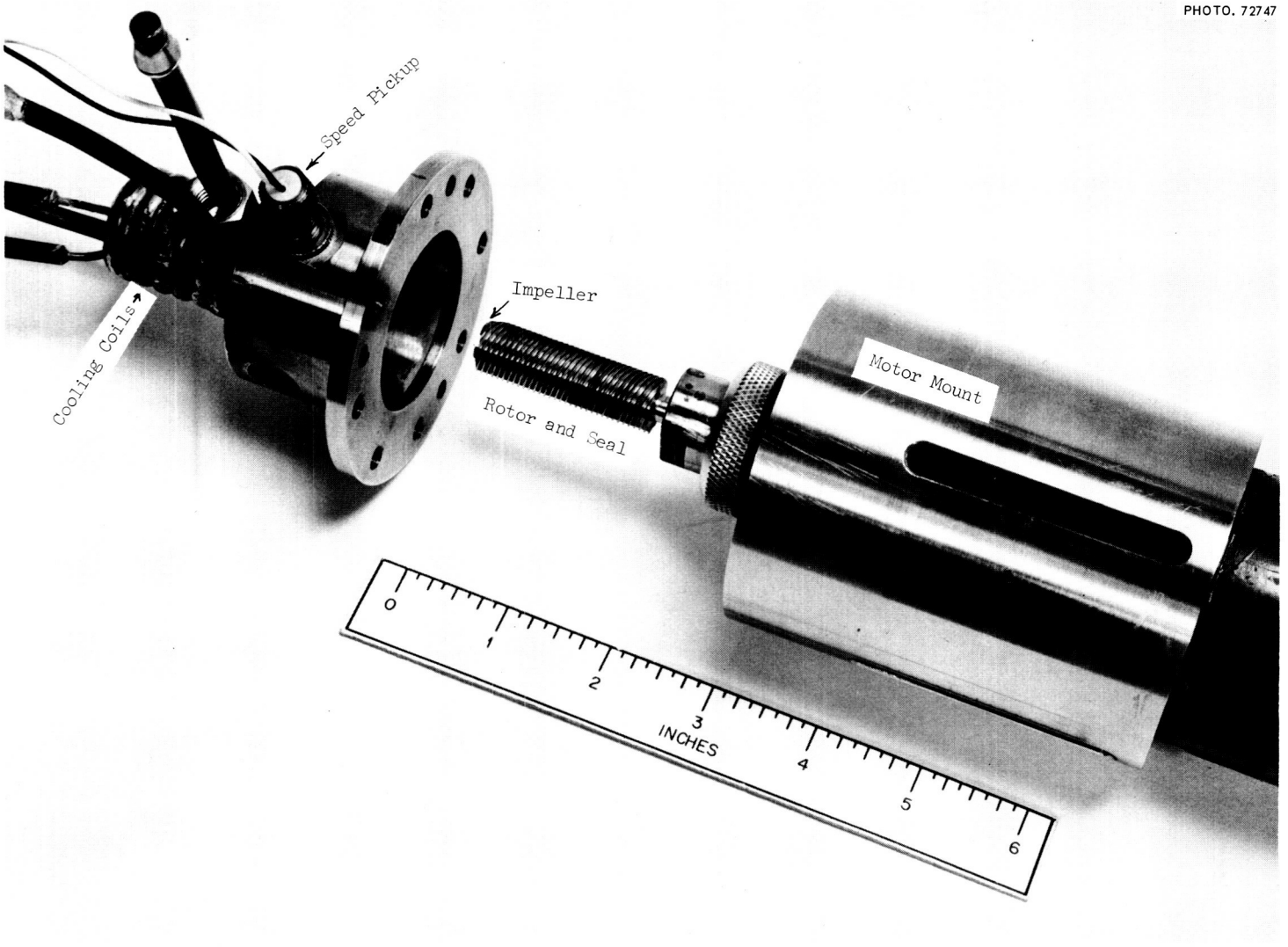

Fig. 8. Photograph of Partially Assembled Test Pump.

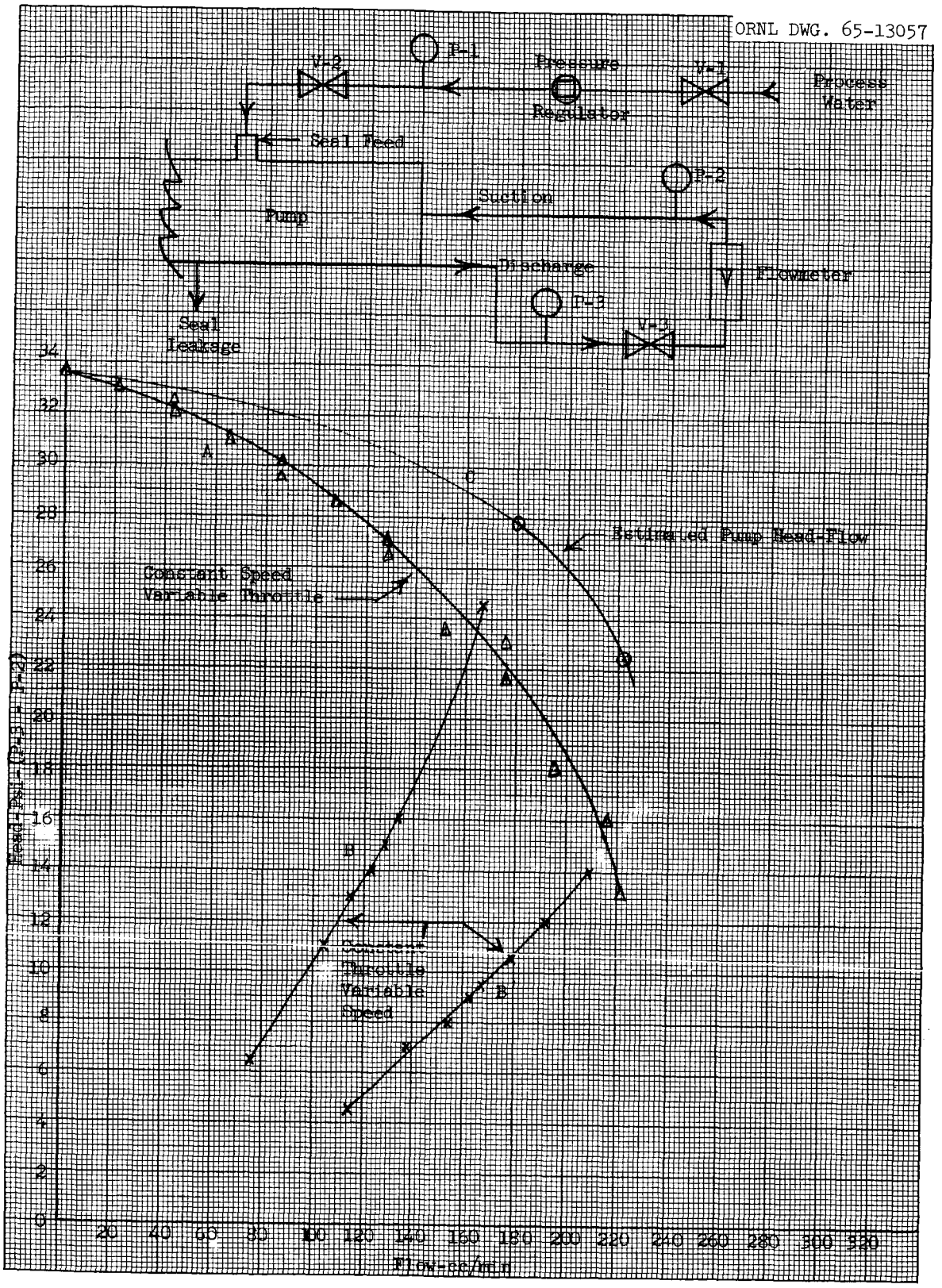

Fig. 9. Head-Flow Results for Test Pump with Water at 20°C.

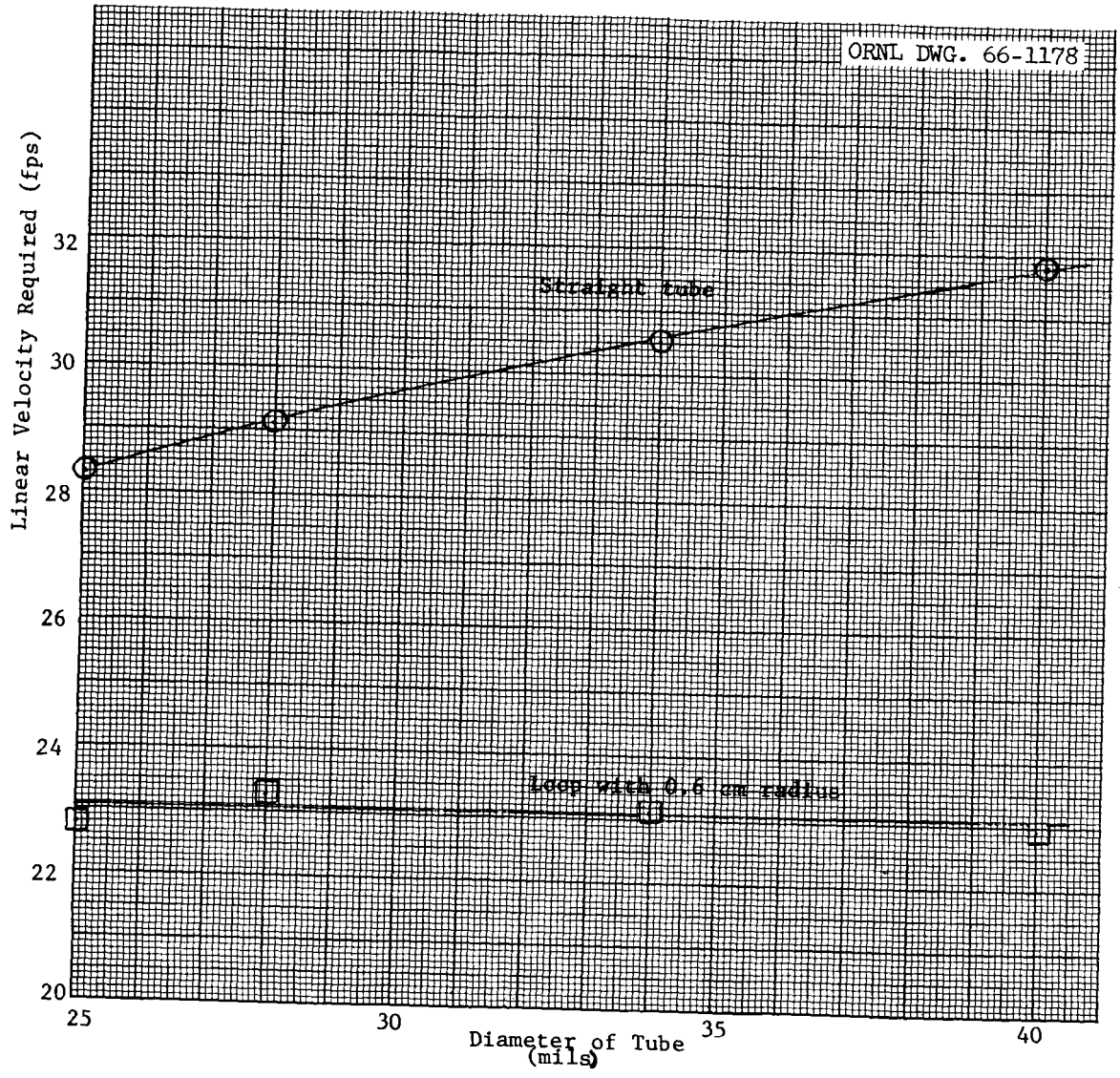

Fig. 10. Calculated Velocity vs Tube Diameter Required to Simulate Fluid Film Conditions in Reactor at 40 fps.

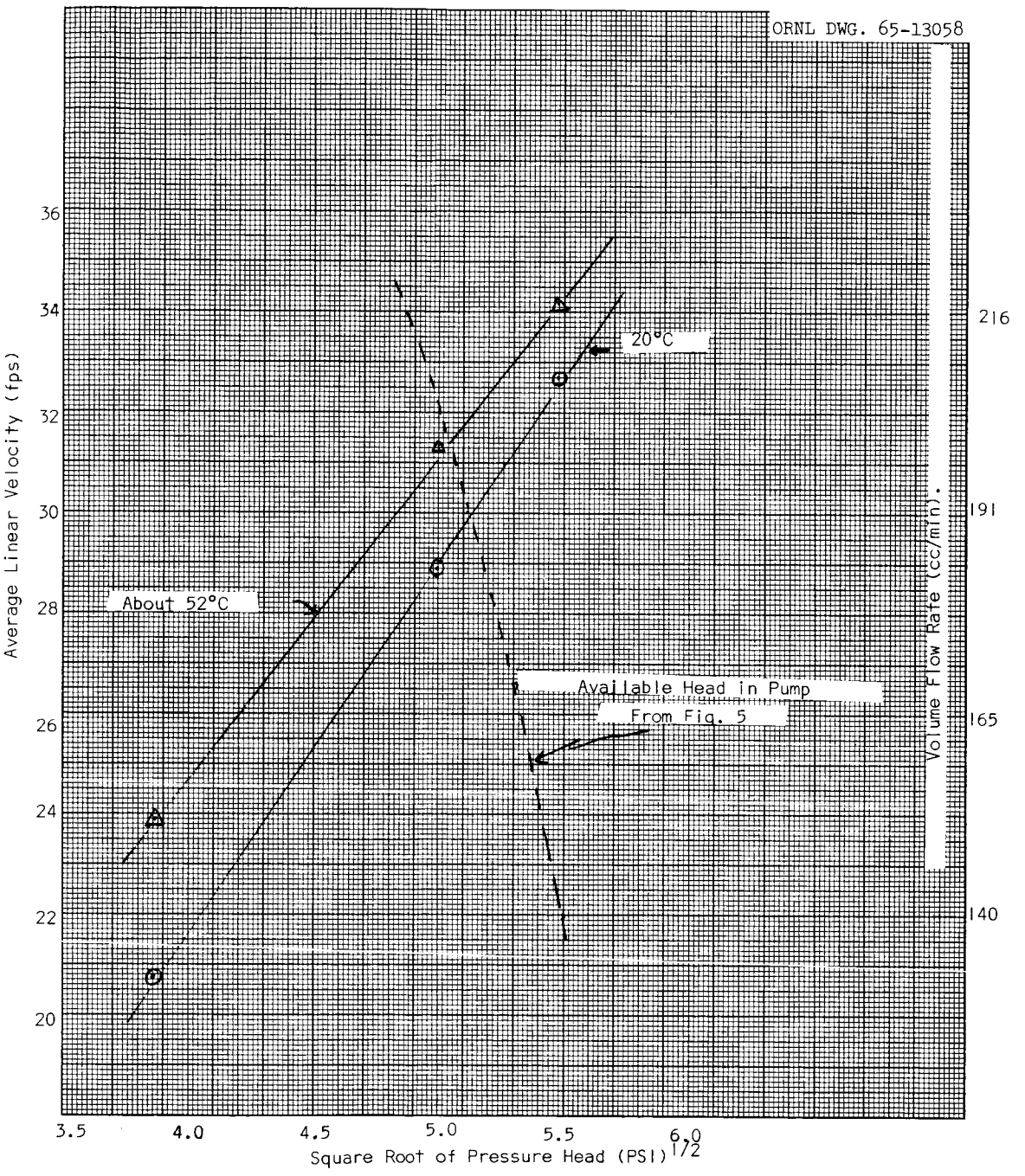

Fig. 11. Pressure Head-Flow Relations for Mock-up of Dynamic System Loop (25.7 mil I.D. Tubing).

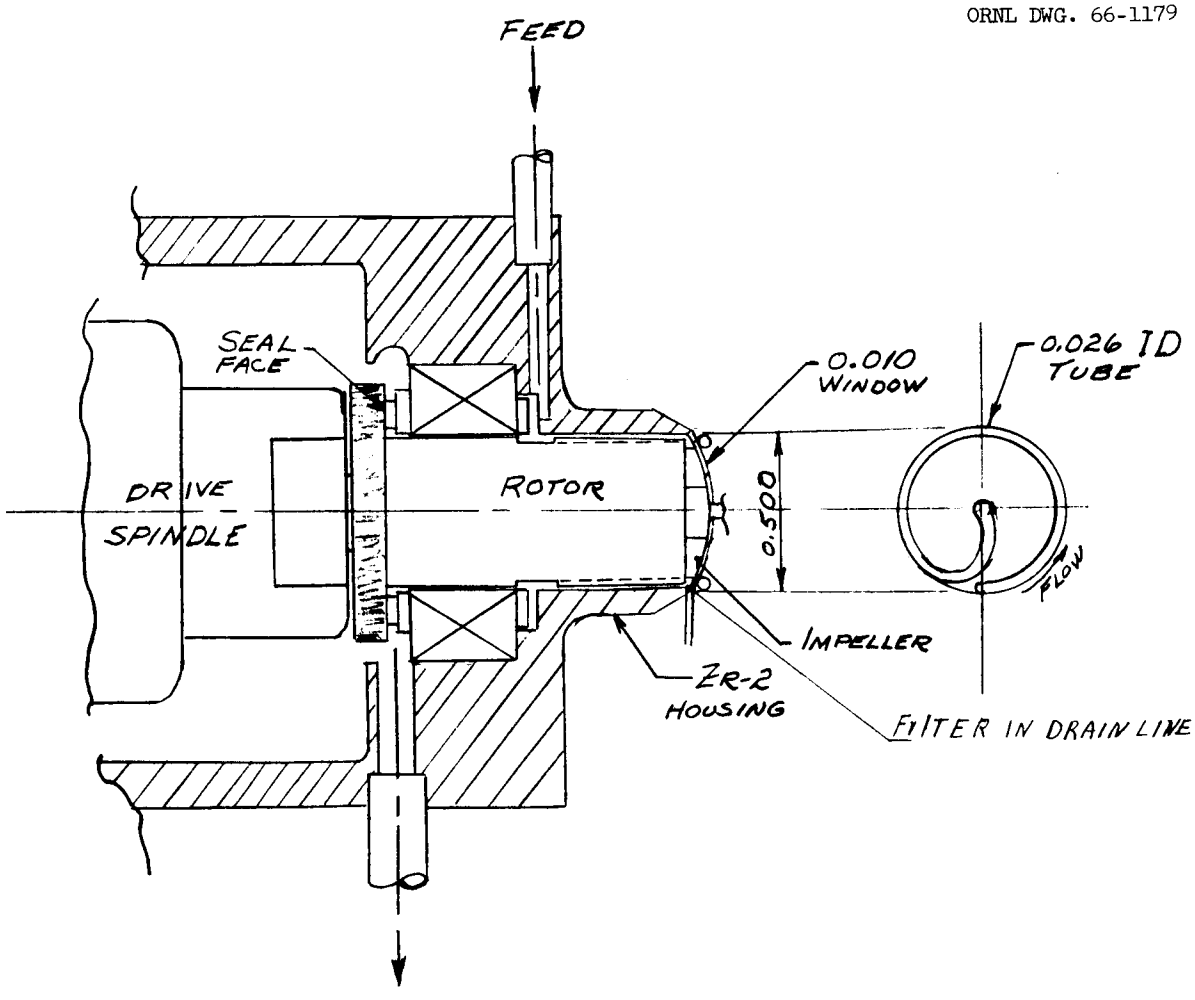

Fig. 12. Proposed Dynamic Cell.

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